Aerosol Size Distribution and Composition from the Twin Otter during CRYSTAL-FACE

Varuntida Varutbangkul¹, Roya Bahreini¹, Jose L. Jimenez², Alice E. Delia², Haflidi H. Jonsson³, Richard C. Flagan¹, and John H. Seinfeld¹

¹ California Institute of Technology / ² University of Colorado, Boulder / ³ Naval Postgraduate School







CRYSTAL-FACE Science Team Meeting February 24-28, 2003

Twin Otter & Relevant Instrumentation

Twin Otter Goals:

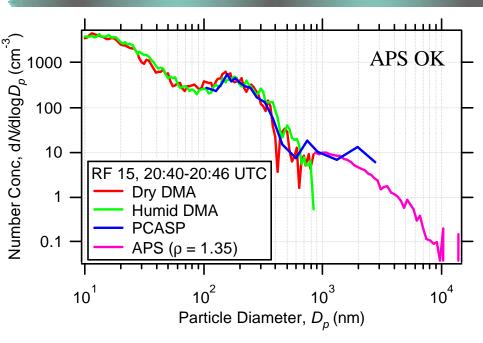
- To characterize aerosols in the boundary layer that are fed into convective systems, possibly participating in cirrus cloud formation
- To measure radiative fluxes under stratiform cirrus anvils.



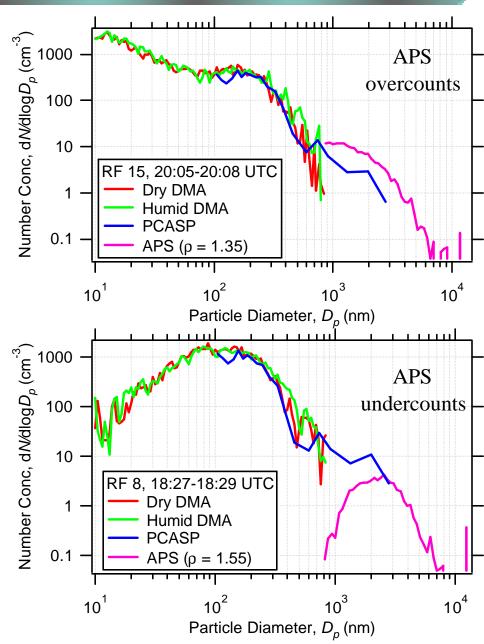
Instrumentation for Measuring Aerosol Size and Composition

| Instrument | Aerosol Characterization | Time Response |
|--|--|---------------|
| Caltech Dual Automated Classified Aerosol Detectors DACAD) • 1 DMA at dry condition (~15% RH) • 1 DMA at humid condition (~70% RH) | Size distribution (10-800 nm) at two relative humidities Aerosol hygroscopicity | 103 s |
| Aerodyne Aerosol Mass Spectrometer (AMS) | Size-resolved (D_{va} = 40-1000 nm) aerosol composition of non-refractory species in time-of-flight mode Integrated aerosol composition in mass spec mode | 60 s |
| PMS Passive Cavity Aerosol Spectrometer Probe (PCASP-100X) | • Size distribution (0.1-3 μm) | 1 s |
| TSI Aerodynamic Particle Sizer (APS) | • Size distribution (0.8-20 µm) | 25 s |

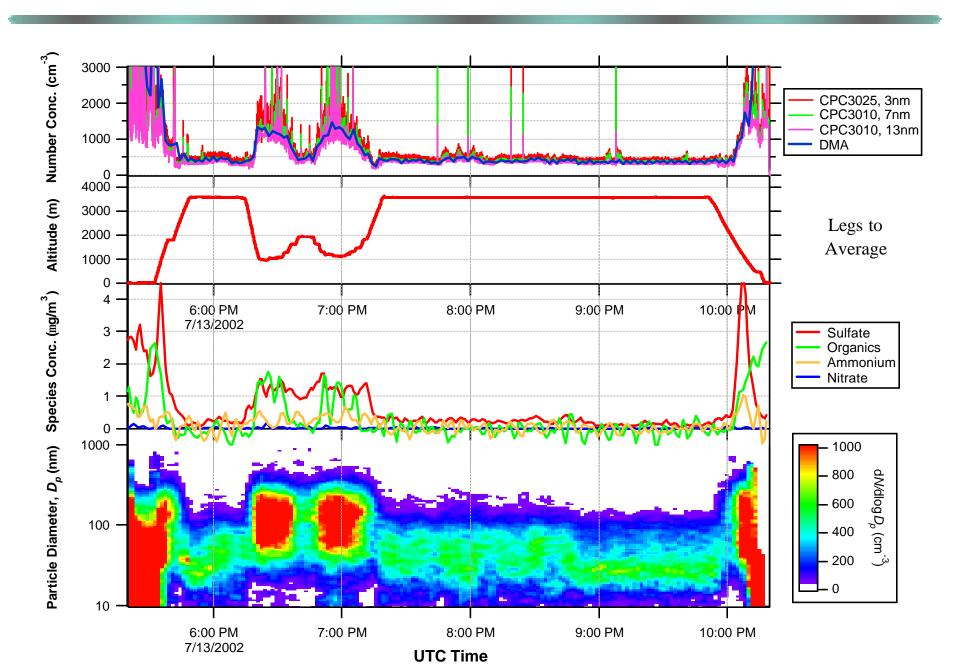
Agreement of Measured Size Distributions



- The overlapping region between the DMA and PCASP (0.1-0.7 μ m) shows good agreement between the two distributions (d*N*/dlog*D*_p within a factor of two of each other at a given size).
- The overlapping region between the PCASP and APS (0.8-3 µm) suggests that the APS often either undercounts or overcounts, but the APS size distribution appears to be reasonable at larger sizes. A super-micron mode is usually not apparent in either measurement.
- The conversion from APS D_a to D_p uses a density estimated from the AMS composition



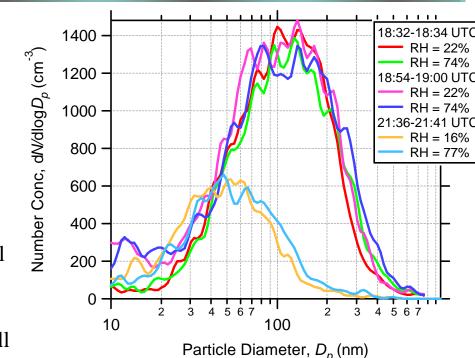
RF 8, 7/13/02: Typical Aerosol



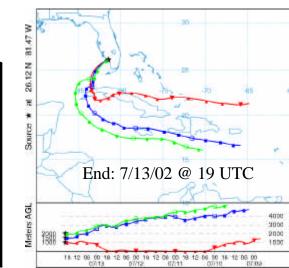
RF 8, 7/13/02: Typical Aerosol

Characteristics of typical aerosol encountered during CRYSTAL-FACE:

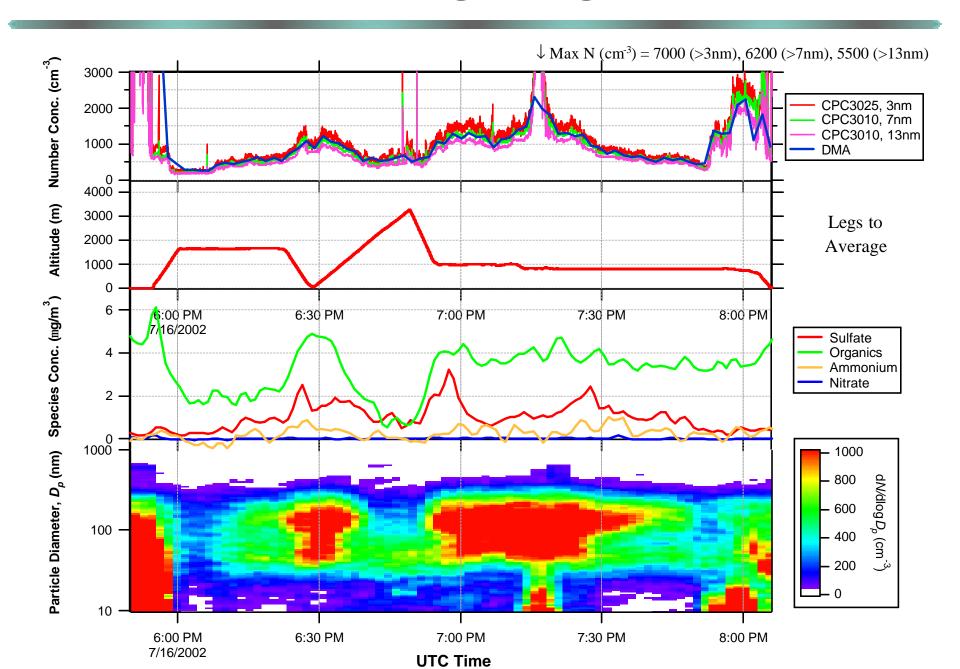
- Low number concentration (< 2000 cm⁻³), relatively homogeneous in number and size distribution within each altitude
- Aerosol composition is primarily sulfate and organics, in roughly 1:1 ratio by mass
- Size distributions show either a weak bimodal shape with a minimum at about 100 nm, or a flat peak at 60-180 nm
- Aerosols at altitudes above 3000 m have small modes between 30-50 nm
- Back trajectory shows marine origin



| RF | UTC Time | Alt (m) | Humid Mode D _p (nm) | Size Distrib. Remarks | Number Conc. (cm ⁻³) | SO_4^{2+} mass $(\mu g/m^3)$ | Organics mass (µg/m³) | Org / SO ₄ | Calc. Density (g/cm³) |
|----|-------------|------------|--------------------------------------|-----------------------------|--|--------------------------------|-----------------------------|--------------------------|-----------------------|
| 8 | 18:32-18:34 | 1267 | ~ 120 | Unimodal | 1058 | 1.31 | 0.78 | 0.6 | 1.51 |
| 8 | 18:54-19:00 | 1137 | ~130 | Unimodal | 1330 | 1.16 | 0.99 | 0.85 | 1.44 |
| 8 | 20:27-20:32 | 3556 | 50 | Unimodal | 453 | 0.34 | < DL | < DL | N/A |
| 8 | 21:36-21:41 | 3557 | 32 | Unimodal | 384 | 0.08 | < DL | < DL | N/A |

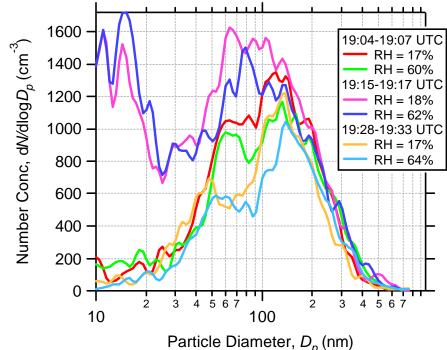


RF 9, 7/16/02: High-Organics Aerosol



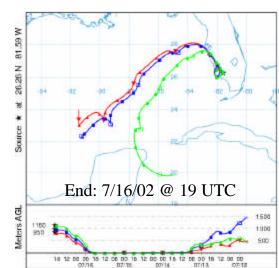
RF 9, 7/16/02 : High-Organics Aerosol

- On 7/16 and 7/18 (RF 9,10,11), the aerosol is largely composed of organic species
- Total number concentration is not significantly elevated over the normal range except for a few spikes in ultrafine aerosol
- AMS sulfate and organic size distributions are similar for D_p between 250 and 500 nm, indicating internally-mixed aerosol in this range
- Back trajectories show influence of continental emissions during the last day of aerosol history

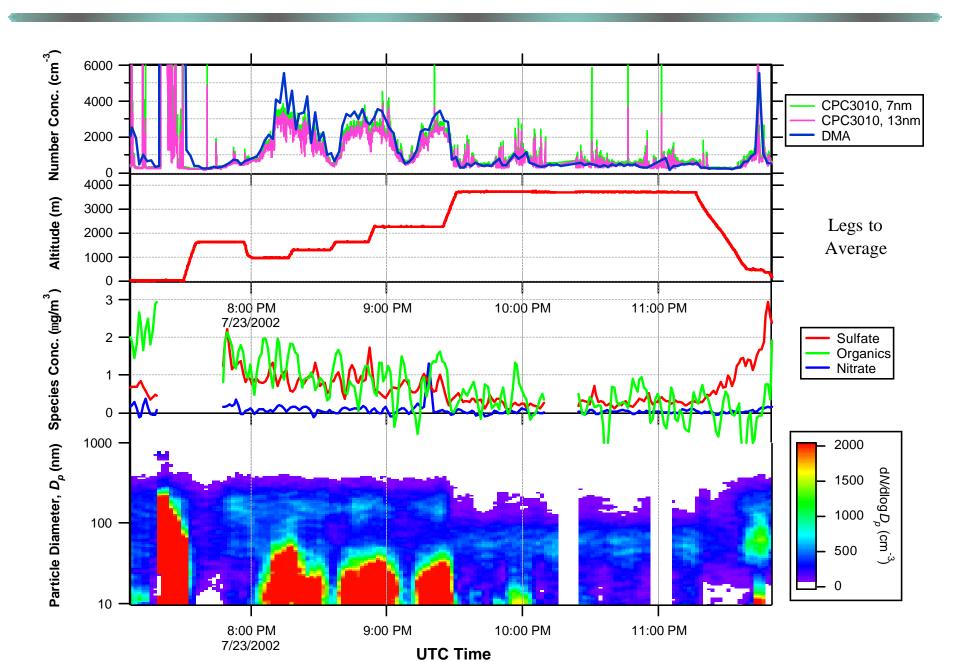


* Total number from CPC 3025 (because of high number of ultrafine aerosol)

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|----|-------------|---------|--------------------------------------|-----------------------------|--|--------------------------------|-----------------------------|--------------------------|-----------------------|
| 9 | 19:04-19:07 | 975 | 65 & 130 | Bimodal | 1063 | 0.71 | 3.54 | 4.99 | 1.09 |
| 9 | 19:15-19:17 | 824 | 15 & 80 | Bimodal | 3732* | 1.04 | 3.94 | 3.79 | 1.13 |
| 9 | 19:28-19:33 | 803 | 60 & 140 | Bimodal | 815 | 1.43 | 3.76 | 2.63 | 1.20 |
| 10 | 16:13-16:16 | 707 | 30 & 125 | Bimodal | 7985 | 2.25 | 7.83 | 3.48 | 1.15 |
| 10 | 16:20-16:21 | 766 | 22 & 120 | Bimodal | 17024 | 2.7 | 8.33 | 3.09 | 1.17 |

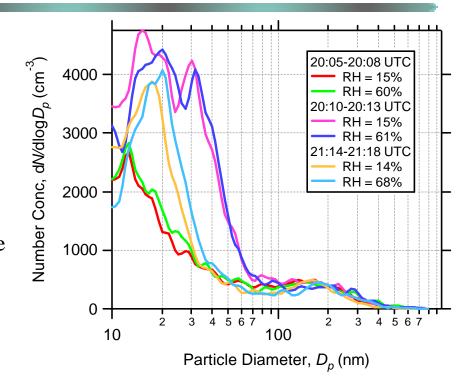


RF 15, 7/23/02 : Fine Aerosol

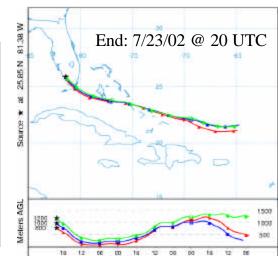


RF 15, 7/23/02 : Fine Aerosol

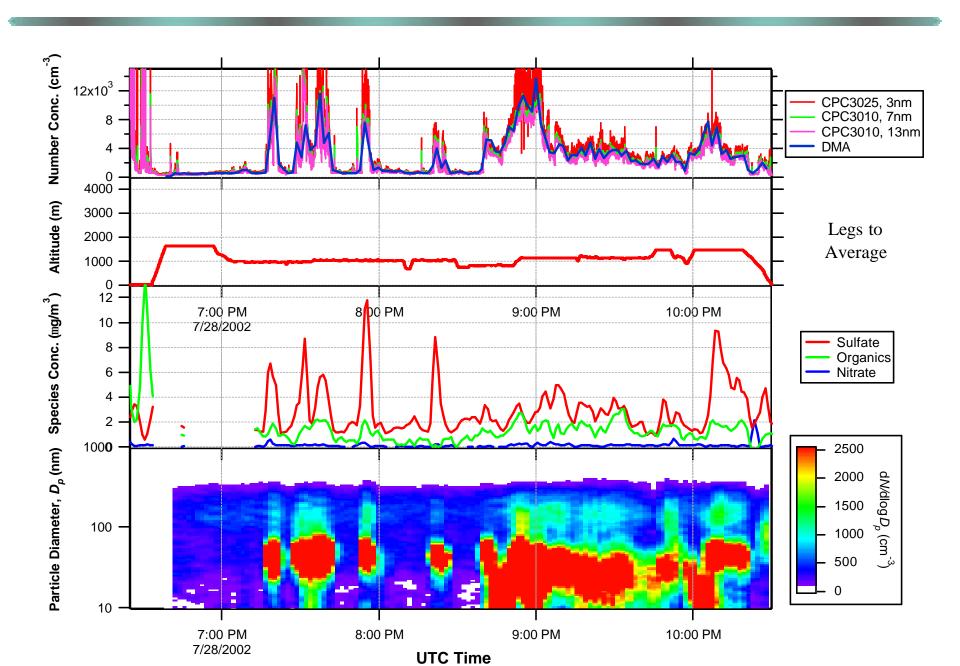
- 7/23 is marked by high concentrations of small nuclei-mode particles below 50 nm, which were observed during cloud-probing at altitudes below 2300 m
- Aerosol profile is also inhomogeneous within an altitude layer.
- Slightly elevated mass ratios of organics to sulfate
- Back trajectory indicates marine origin which could implicate relatively freshly-nucleated natural particles



| RF | UTC Time | Alt (m) | Humid Mode D _p (nm) | Size Distrib. Remarks | Number Conc. (cm ⁻³) | SO_4^{2+} mass $(\mu g/m^3)$ | Organics mass (µg/m³) | Org / SO ₄ | Calc. Density (g/cm³) |
|----|-------------|---------|--------------------------------------|-----------------------------|--|--------------------------------|-----------------------------|--------------------------|-----------------------|
| 15 | 20:05:20-08 | 971 | 13 & 150 | Bimodal w/ ultrafine | 1974 | 0.79 | 2.1 | 0.73 | 1.20 |
| 15 | 20:10-20:13 | 971 | 20 & 30 | Fine modes dominate | 4008 | 0.88 | 1.61 | 0.65 | 1.27 |
| 15 | 21:14-21:18 | 2245 | 20 & 170 | Bimodal sharp peak | 3332 | 1.11 | 1.31 | 0.54 | 1.37 |

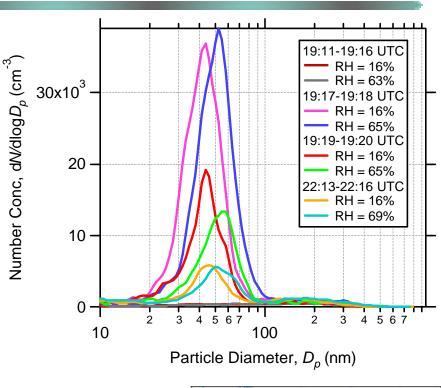


RF 18, 7/28/02: Non-homogeneous Air Mass

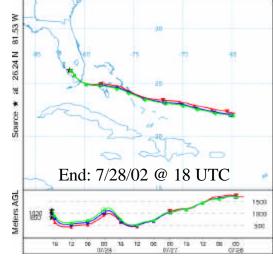


RF 18, 7/28/02: Non-homogeneous Air Mass

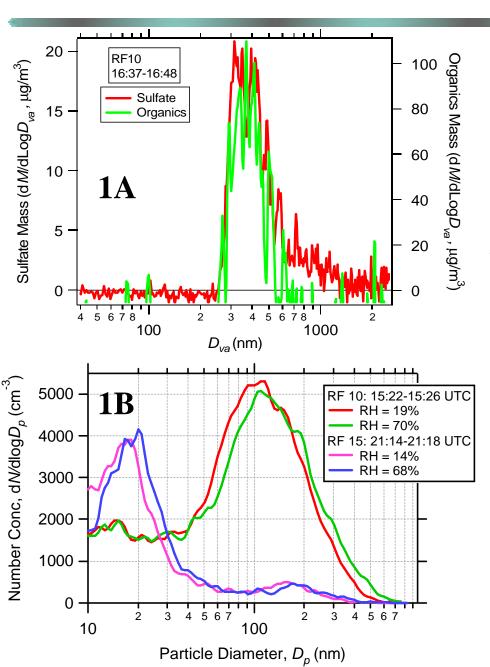
- 7/28 is marked by uncommonly inhomogeneous air mass, characterized by frequent spikes in number concentration, mainly of nuclei-mode and ultrafine aerosol, even at a constant altitude
- Inhomogeneity indicates on-going mixing process. This is counter-intuitive because back trajectories show a very homogeneous history of the air mass, both in terms of altitude and insensitivity to model start time.
- The inhomogeneity may be due to the entrainment or formation of fresh particles in close proximity to the sampling location
- The time scale of change in the number concentration is sometimes too fast for a DMA scan to completely capture the time variation in size distribution.



| RF | UTC Time | Alt (m) | Humid Mode D _p (nm) | Size Distrib. Remarks | Number Conc. (cm ⁻³) | SO ₄ ²⁺ mass (μg/m ³) | Organics mass (µg/m³) | Org / SO ₄ | Calc. Density (g/cm³) |
|----|-------------|------------|--------------------------------------|-----------------------------|--|---|-----------------------------|--------------------------|-----------------------|
| 18 | 19:11-19:16 | 958 | ~65 & 170 | weakly bimodal | 588 | 1.38 | 1.22 | 0.88 | 1.43 |
| 18 | 19:17-19:18 | 964 | 52 | Fine peak growing | 4461 | 7.6 | 1.17 | 0.15 | 1.68 |
| 18 | 19:19-19:20 | 975 | 55 | Fine peak dominates | 11222 | 5.58 | 1.88 | 0.34 | 1.6 |
| 18 | 22:13-22:16 | 1466 | 50 & 180 | Bimodal | 2808 | 5.22 | 1.31 | 0.25 | 1.64 |

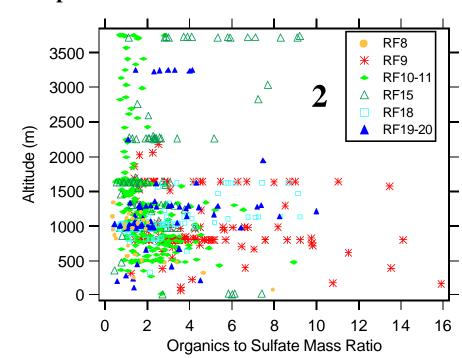


General Observations



1. Evidence of internal mixing

- A. AMS: Similar mass distributions of sulfate and organics, at least for the D_p range of 250-500 nm
- B. DMA: Similar growth of particles below 500 nm when exposed to the same RH (note that number concentrations above 500 nm are small; thus, hygroscopic growth becomes difficult to determine)
- 2. Higher organic-to-sulfate mass ratios are generally found at lower altitudes, with the exception of RF 15



Summary & Acknowledgements

Summary:

- Various types of air masses were sampled in the Twin Otter flights
- The time scale of change in the aerosol number concentration depends on the size of interest: the fine nuclei mode aerosol ($D_p < 100 \text{ nm}$) varies with a much shorter time scale than that of the accumulation mode ($D_p > 100 \text{ nm}$). Spikes of high aerosol number often occur in the ultrafine size range.
- Above the altitude of ~ 3000 m, particles becomes sparse and are generally uniform in number and size, with a mode between 30-50 nm
- Aerosol below 500 nm is predominantly internally-mixed, as evidenced by:
 - Composition that is generally uniform with size
 - Similar growth behavior with RH

Acknowledgements:

Funding:

- NASA
- Office of Naval Research

Analysis Tools:

- James Allan (UMIST, AMS Analysis Software)
- NOAA Air Resources Laboratory (ARL) for provision of the HYSPLIT transport and dispersion model